ESTIMATION OF MAXIMUM PERMISSIBLE STEP LOSSES IN P&T PROCESSING

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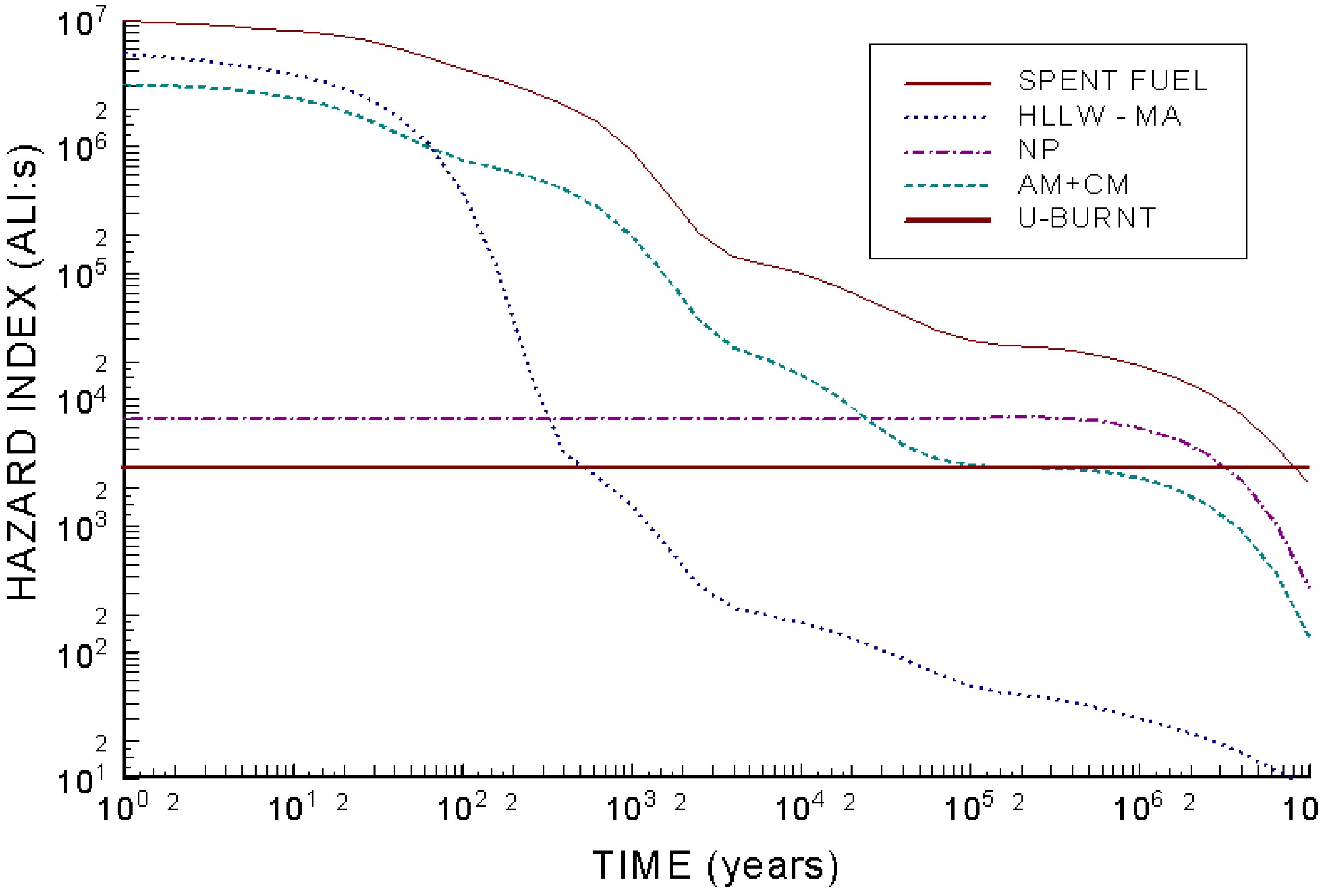
MAXIMUM TOTAL LOSSES

As a basis for the calculation of permissible step losses we need to define what is the permissible total loss to waste as a fraction of the mass of each element containing radioisotopes.

This can in the end only be made by lawmakers.

However, in order to illustrate the mathematical methods we will base the maximum total loss fraction of any element on a potential hazard index for the final waste.

An Example of a Potential Hazard Index



ESTIMATED MAXIMUM TOTAL LOSSES

When the potential hazard index is smaller than that of the potential hazard of the uranium burnt in the reactor when producing the radioactive wastes, we can argue that the total hazard in nature remains unchanged.

In case of Np a reduction of the total amount in the waste by a factor of >2.5 results in a potential hazard from that element alone which crosses our reference line at about 400 years.

For the other minor actinides, Am and Cm, their amount must be reduced by a factor of >124 to reach our reference line after 400 years.

Both requirements should to be within reach for P&T.

FLOWSHEETS FOR P&T OF MINOR ACTINIDES

There are in principle only two alternative flow-sheets:

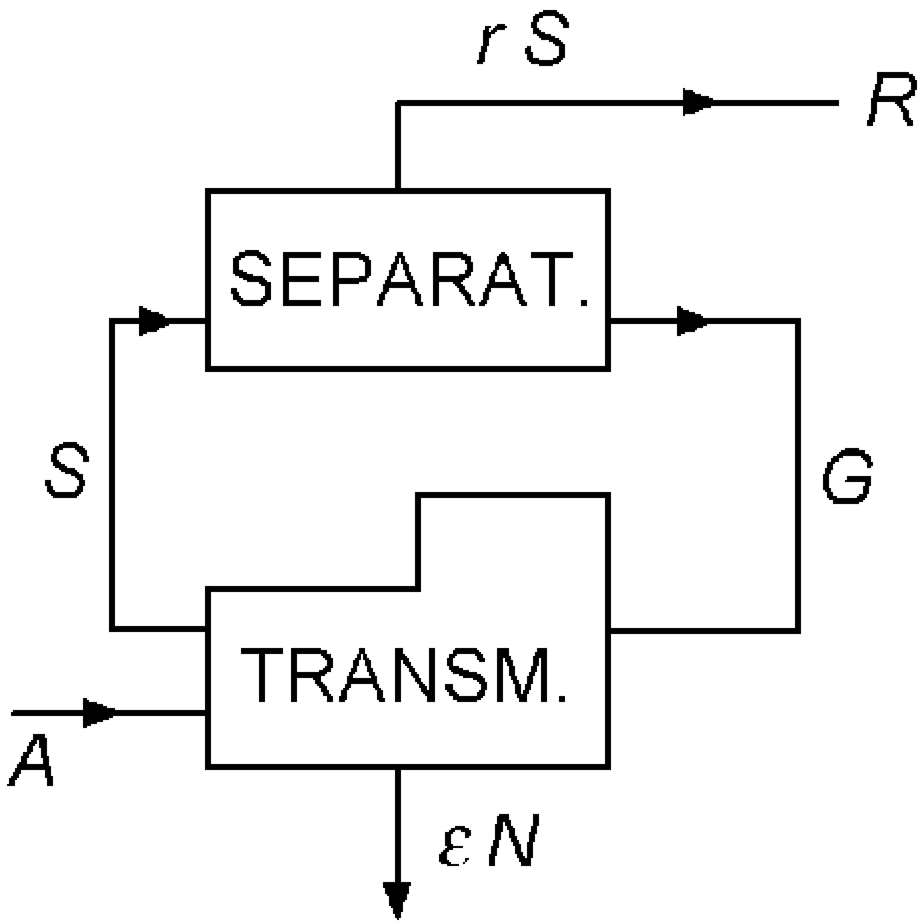
1. On-line continuous processing and purification of a continuous target stream This requires some kind of liquid fuel reactor or, perhaps, a pebble-bed reactor.

2. Batch-wise off-line processing of irradiated target material. This is the classical way to operate the nuclear fuel cycle and target/fuel can i principle be of any type.

I will begin by showing how losses from a continuous system can be calculated.

LOSSES FROM CONTINUOUS P&T

A simplified flow-sheet for continuous operation:



A = Rate of addition of target atoms to transmuter

N = Number of atoms in transmuter

S = Transfer rate to separation process

A = ε N + r S; where ε is transmutation rate constant and r is fraction of S lost to waste.

C = N / V; where V is the volume in the transmuter

v = k V = volume flow-rate to separation process

S = v C = transfer rate to separation

R = r S = loss-rate to waste

$\varepsilon N = \phi_{avg} \sigma_{avg} N$; thus $\varepsilon = \phi_{avg} \sigma_{avg}$

Q = A/R = reduction factor from current total loss fraction

Some simple arithmetic results in the following expression for r: $r = \phi_{avg} \sigma_{avg} / [k (Q-1)]$;

($\phi_{avg} \sigma_{avg}$ is of the order of 10^{-8} for fast neutrons.)

Application to Np assuming a Q-value of 2.5

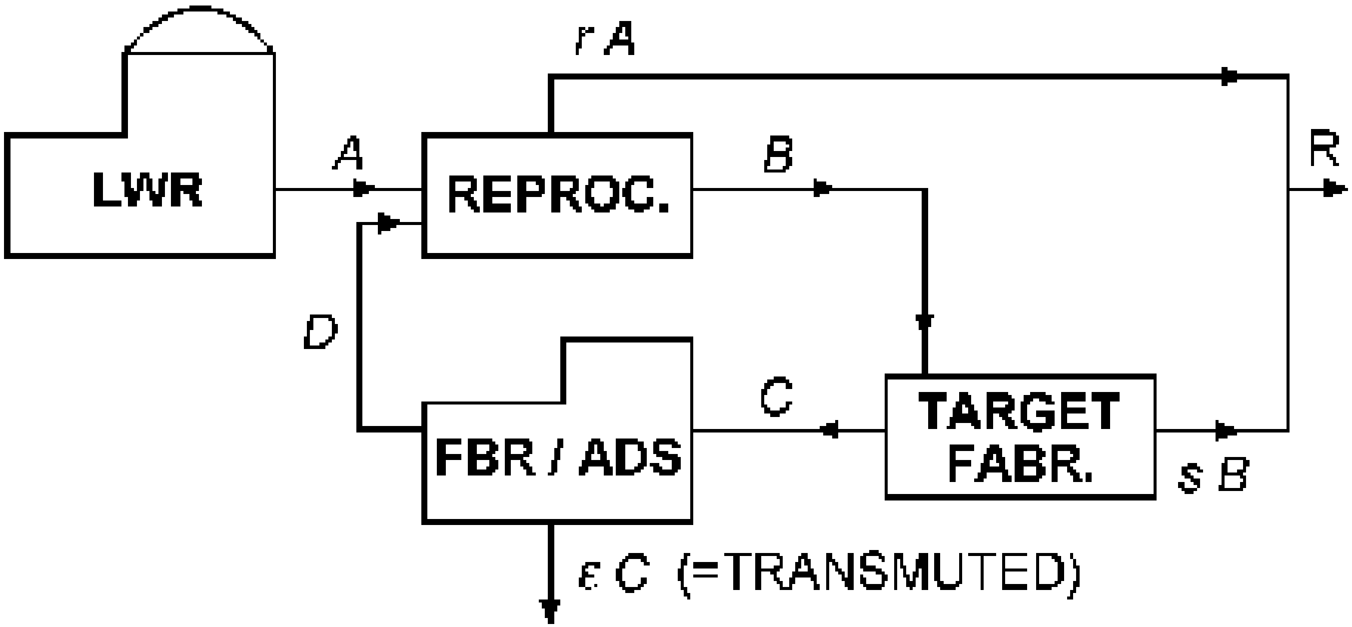
σ_{avg} = ~ 2 b = 2x10^{-28} m^2 (in fast reactor or ADS system)
ϕ_{avg} = ~ 1x10^{19} n m^2 s^{-1}
Q = 2.5
Assume that 1 transmuter volume is purified per day
k = (1/86400) s^{-1} = 1.16x10^{-5} s^{-1}

Then
r = (1x10^{19} x 2x10^{-28})/[1.16x10^{-5} x (2.5-1)] = 1.15x10^{-4} = 0.0115 % of separated amount of Np.

I we instead assume that 1 transmuter volume is purified per week we obtain a permissible loss-fraction, r, of 0.081 %

LOSSES FROM BATCH-WISE P&T

A simplified flow-sheet for batch-wise operation:



The calculation trick is to follow a given amount around and around until all is either transmuted or lost to wastes.

Symbols, definitions and calculations:

A = incoming amount in a single batch, first cycle

B = (1 - r) A = amount remaining after reprocessing

C = (1 - s) B = amount entering next T-cycle, but

C = (1 - r) (1 - s) A

D = (1 - ε) C = amount NOT transmuted

D = (1 - r)(1 - s) (1 - ε) A

Now D is the amount recirculating a second time. This for the 2:nd cycle we have

B' = (1 - r) D = $(1 - r)^2(1 - s)(1 - e)$ A

C' = (1 - s) B' = $(1 - r)^2(1 - s)^2$ A

D' = (1 - ε) C' = $(1 - r)^2(1 - s)^2(1 - \varepsilon)^2$ A

Etc. cycle after cycle. Finally after an infinite number of cycles we have:

D = $(1 - r)^\infty (1 - s)^\infty (1 - \varepsilon)^\infty$ A

The total amount lost to waste, R, can now be calculated by summing all losses from processing and target fabrication during an infinite number of cycles. Hence:

$$R = A\,(r + r\,(1 - r)(1 - s)(1 - \varepsilon) + r\,(1 - r)^2(1 - s)^2(1 - \varepsilon\,)^2 + \ldots) +$$
$$+ A\,(\,s\,(1 - r) + s\,(1 - r)^2\,(1 - s)(1 - \varepsilon) + s\,(1 - r)^3\,(1 - s)^2\;1 - \varepsilon)\;^2 + \ldots)$$

After n cycles we then have (sums are for m = 0 to n):

$$R = A\,r\,\Sigma((1 - r)^m\,(1 - s)^m\,(1 - \varepsilon)^m + A\,s\,(1 - r)\,\Sigma(1 - r)^m\,(1 - s)^m\,(1 - \varepsilon)^m$$

This expression can now be simplified and written as:

$$R = A\,(r + s - s\,r)\,\Sigma(1 - r)^m\,(1 - s)^m\,(1 - \varepsilon)^m$$

However, all values within parenthesis with exponent m are smaller than 1. Thus we can use the rules for sums of an infinite power series.

Thus after an infinite number of cycles we have:

$$R = A\,(r + s - s\,r)\,/(r + s + \varepsilon - r\,s - r\,\varepsilon - s\,\varepsilon + r\,s\,\varepsilon)$$

Now we can write an expression for the total loss fraction as:

$$R / A = (r + s - s\,r)\,/(r + s + \varepsilon - r\,s - r\,\varepsilon - s\,\varepsilon + r\,s\,\varepsilon) = 1 / Q$$

Application to Np assuming a required Q-value of 2.5

$s = 0.0001$ (from MOX-fuel experience and
$\varepsilon = 0.05$ (estimated from SUPERFACT experiments)
Hence:
$r = (Q\,s - s - \varepsilon + s\,\varepsilon)/(1 - Q + Q\,s - s - \varepsilon + s\,\varepsilon) = 0.032 =$
3.2 % relative loss-fraction per cycle could be tolerated

CONCLUSIONS

1. Methods have been developed which permit the calculation of permissible step-losses in either continuous or batch-wise P&T operations from given values of the total acceptable loss fraction found in radioactive waste

2. In the case of Np, and based on a relative hazard calculation, it was found that the acceptable step losses are much smaller in any continuous process than in a discontinuous process. This can also be intuitively understood from a consideration of entropy changes in the two alternatives.

3. The equations derived permits us to compare the requirements on both the P and the T part of any P&T process for reduction of the amount of long-lived nuclides in a repository by a given factor.

Acknowledgment

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